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Waste-to-Energy biofuel production potential for selected feedstocks in the conterminous United States

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ABSTRACT

Waste-to-Energy (WtE) technologies offer the promise of diverting organic wastes, including wastewater sludge, livestock waste, and food waste, for beneficial energy use while reducing the quantities of waste that are disposed or released to the environment. To ensure economic and environmental viability of WtE feedstocks, it is critical to gain an understanding of the spatial and temporal variability of waste production. Detailed information about waste characteristics, capture/diversion, transport requirements, available conversion technologies, and overall energy conversion efficiency is also required. Building on the development of a comprehensive WtE feedstock database that includes municipal wastewater sludge; animal manure; food processing waste; and fats, oils, and grease for the conterminous United States, we conducted a detailed analysis of the wastes' potential for biofuel production on a site-specific basis. Our analysis indicates that with conversion by hydrothermal liquefaction, these wastes have the potential to produce up to 22.3 GL/y (5.9 Bgal/y) of a biocrude oil intermediate that can be upgraded and refined into a variety of liquid fuels, in particular renewable diesel and aviation kerosene. Conversion to aviation kerosene can potentially meet 23.9% of current U.S. demand.

1. Introduction

Waste-to-Energy (WtE) technologies offer the promise of providing a synergistic relationship between industry and various levels of government to divert organic wastes such as wastewater sludge, agricultural and livestock waste, food waste, and municipal solid waste for beneficial energy use, while reducing the quantity of waste disposed and/or released to the environment. The approach is to make beneficial use of waste resources in a manner that 1) potentially eliminates, or at least significantly reduces adverse effects on public health, safety, welfare, and/or the environment; 2) contributes to sustainability factors; and 3) provides a net positive energy outcome. An important consideration in the WtE landscape is the waste management hierarchy (Fig. 1) that generally depicts a prioritization in the waste management process, wherein the focus is to minimize and divert waste, and then, only as a final option, dispose of it. This paper is centered on the beneficial use of waste resources after efforts have been made to reduce and avoid waste, and reuse, recycle, and compost waste where possible. The top of the hierarchy (waste reduction/avoidance) identifies the most preferred and sustainable option, and the least preferred and last resort option is waste disposal/release. The hierarchy is general, and a given feedstock and decision making around that feedstock may not always fit this structure. Some examples include 1) the conversion of waste to a biofuel may provide a higher value use than recycling/composting, and 2) reuse of wastewater sludge requires additional treatments to produce Class A/B biosolids, and thus a direct energy

Abbreviations: BETO, U.S. Department of Energy's Bioenergy Technology Office; Bgal, billion gallons; CAFO, concentrated animal feeding operation; EERE, U.S. Department of Energy's Office of Energy Efficiency and Renewable Energy; EPA, U.S. Environmental Protection Agency; FOG, fats, oils and grease; GL, gigaliters; HHV, higher heating value; HTL, hydrothermal liquefaction; IIC, industrial, institutional, and commercial; Mg, megagrams; Mgal, million gallons; MJ, megajoules; ML, megaliters; MT, million U.S. short tons; PNNL, Pacific Northwest National Laboratory; POTW, publicly owned treatment works; Tg, teragram; US, United States of America; VSS, volatile suspended solids; WtE, Waste-to-Energy

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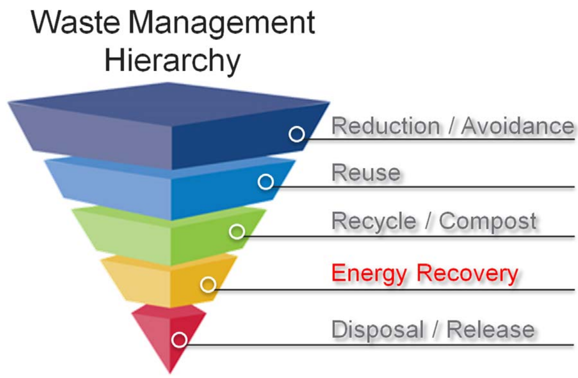

Fig. 1. Waste management hierarchy defining prioritization of handling waste, where reduction/avoidance is the most preferred and sustainable, whereas disposal/release is the least preferred and sustainable option.

recovery pathway may be more efficient.

These organic wastes provide a reservoir of carbon resources for energy production that otherwise represent mounting challenges associated with greenhouse gas emissions, pollutant reduction, and various measures of sustainability. In addition, trends toward more concentrated operations realize economies of scale—for example, fewer but larger solid waste landfills and fewer but larger concentrated animal feeding operations (CAFOs). Thus, a synergistic relationship among waste handlers offers an economic opportunity for converting waste liabilities into revenue streams or cost-neutral endeavors. Emerging trends toward decentralized fuel production also offer the opportunity to colocate properly scaled feedstock conversion facilities with a blend of compatible feedstocks. A critical challenge to ensuring the economic and environmental viability of WtE feedstocks is gaining an understanding of the spatial and temporal variability of waste production, characteristics, capture/diversion and preprocessing methods, transport requirements, available conversion technologies, and the overall energy return on investment. This understanding can then lead to more accurate estimates of energy and co-product production potential. Associated demand for other resources such as water, land, critical infrastructure, and additional opportunities for co-product generation (e.g., fertilizers) can also be evaluated.

The objective of this paper is to provide a foundation for a robust WtE industry that can capitalize on underutilized organic wastes for biofuels production in the conterminous United States. To support this objective, a comprehensive spatially enabled WtE feedstock database was developed that includes municipal wastewater sludge, animal manure, food processing waste, and fats, oils, and grease (FOG) [1,2]. This database development enabled us to carry out a detailed site- and feedstock-specific resource assessment to assess the biofuel production potential. Given the variability of these feedstocks and potential implications for downstream biorefinery design and operation, robustness in the energy conversion pathway (characterized by diversity, adaptability, and efficiency) is imperative. For the purposes of this initial assessment, we assumed hydrothermal liquefaction (HTL) to be the target pathway, because, as a conversion technology for wet biomass, it is rapidly approaching market readiness [3–6]. Results from these analyses will enable the U.S. Department of Energy and its stakeholders to accurately evaluate the scale and viability of WtE potential contributions to the Bioenergy Technologies Office Multiyear Program Plan target dry-weights of 245 Tg/y by 2017 and 285 Tg/y by 2022 [7].

This paper is organized as follows. Section 2 summarizes a resource assessment of selected WtE feedstocks for the conterminous United States including production potential and general characteristics. Section 3 provides an overview of HTL as a representative conversion pathway for these feedstocks to produce a biocrude oil intermediate. In Section 4, a reduced-form HTL conversion model, used to estimate the biocrude oil production potential of each feedstock, is described. Results of the biocrude oil production assessment, including the spatial distribution of the feedstocks, are summarized and discussed in Section 5. Section 6 highlights the key conclusions and recommended next steps for this research.

2. Selected WtE resources overview

2.1. Wastewater sludge

Management and disposal of municipal wastewater sludge is a significant challenge throughout the United States and can be of considerable expense to treat and/or dispose given its significant volume, high water content, and pollutant concentrations (e.g., pathogens, heavy metals, pharmaceuticals, persistent organics, etc.). Generally, the methods and practices for wastewater treatment and sludge management in the United States are founded in engineering traditions dating to the early 20th century, and are primarily driven by considerations of function, safety, and cost-benefit analysis. However, looking to the future with a sustainability and beneficial-use perspective, wastewater can be viewed as a renewable resource from which we can recover water, nutrients, and energy produced from the high organic content in the waste stream. Maximizing water reclamation and unconstrained reuse can be an important asset in water-stressed areas.[1] As an example of energy recovery, anaerobic digestion has been practiced for decades to generate methane for onsite heat and/or electricity generation, and some facilities have achieved or neared a net-zero energy footprint. At many facilities, the production of biosolids for fertilizer/soil amendments is a beneficial use of the sludge waste; however, social concerns about this practice have increased with regard to heavy metals and pharmaceutical compounds being introduced to soils used for crop production [8]. As such, current wastewater treatment practices are believed to predominantly have a negative effect on local/regional water, energy, and material sustainability [9]. Additionally, there is an increasing frequency of cases throughout the United States where summertime algal blooms severely affect freshwater resources (municipal water, irrigation water, recreation, wildlife, etc.). In part, this results from long-term accumulated and excess available nitrogen and phosphorous within the aquatic environment in combination with warm water bodies (i.e., shallower water depths; higher summertime temperatures) that provide favorable growing conditions for varying types of algae and cyanobacteria [10–13]. In the future, areas more prone to algal blooms may require the diversion of treated wastewater streams or be subject to increased regulation of nutrient concentrations released in treated wastewater. Addressing such diversion or regulatory needs can in part be solved by the beneficial use of sludge, including HTL processing for biocrude oil and nutrient recovery [14].

Within a given publicly owned treatment works (POTWs) design, a number of unit processes can be implemented for sludge processing, depending on the plant design and operation, plant objectives, and characteristics of the waste stream being processed [15,16]. The principal sources of sludge considered for WtE in this study are primary and secondary treated waste. Primary treatment involves the initial clarification or settling of suspended solids (i.e., primary sedimentation). Chemical flocculants are often used to increase the efficiency (time to settle and total solids) of solids settling. Primary treatment consists of concentrating organic solids and inorganic fines to 2–7% concentration, where 40–70% of total suspended solids are captured with an approximate solids production of 0.1–0.3 kg/m^3 of wastewater [17,18]. Volatile suspended solids (VSS) concentration generally ranges from 60% to 85%.

Secondary treatment is focused on biological treatment and involves a combination of aeration, exposure to microbes, and secondary clarification through additional solids settling (i.e., secondary

[1] One example is the Pure Water San Diego project: http://PureWaterSD.org.

sedimentation). This treatment focuses on the dissolved organic matter that was not captured during the primary treatment phase. The outcome of this treatment stage is waste-activated sludge, the bulk of which goes to sludge generation/removal and the remaining portion is returned to the aeration and clarification stage (return activated sludge). During this phase, solids are concentrated to 0.5–1.5% with VSS concentrations between 70% and 80%. Some secondary treatment systems also include a trickling filter that produces a sludge that is 4–5% solids and 45–70% VSSs. In general, the secondary treatment increases the total solids removal to 85%. The output of the waste-activated treatment goes through a mechanical thickening/dewatering stage that increases the solids concentration of the sludge to somewhere between 15% and 40%. The primary and secondary treatment sludge can also be blended into a single waste stream that can be dewatered to an appropriate concentration.

Based on facility data cataloged in the U.S. Environmental Protection Agency's (EPA) 2008 and 2012 EPA Clean Water Needs Surveys and the EPA Integrated Compliance Information System National Pollutant Discharge Elimination System, Seiple et al. [1] inventoried 15,014 POTWs throughout the conterminous United States, Alaska, Hawaii, and Puerto Rico, which serve approximately 76% of the total U.S. population. Seiple et al. [1] further estimated that in total these POTWs treat on average 130.5 GL/d (34.5 Bgal/d) of wastewater and generate 12.56 Tg/y (13.84 MT/y) of dry-weight solids, of which 7.43 Tg/y (8.19 MT/y) are primary treated sludge and 5.13 Tg/y (5.65 MT/y) are secondary treatment sludge. The number of POTWs and daily flow from all facilities is distributed by the facility size in Fig. 2 [1]. The majority of the POTWs are sized ≤ 5 ML/d (1.3 Mgal/d), accounting for 42% of all treatment plants; however, the fraction of daily flow for these same size plants only accounts for 1% of all influent to be treated. POTWs sized at 50–100 ML/d (13.2–26.4 Mgal/d) handle the majority of influent, though these only represent 3% of all POTWs. A breakdown and summary of the national wastewater sludge resource is presented in Table 1.

2.2. Animal manure

Animal manure is used as an inexpensive fertilizer or additive to improve soil quality, and currently, an estimated 8% of all U.S. cropland is fertilized in this manner [19]. Factors driving the use of manure are the agronomic needs of crops and transport cost, which limit the haul distances, thus creating close links between types of livestock and crops. Corn is planted on ~ 25% of U.S. cropland, and accounts for over half of the land using manure [20]. The bulk of the manure applied to corn is generated from nearby dairy and swine operations. Manure from poultry and cattle feedlot operations is drier, thus less costly to transport, and often shipped to other operations—for example, for use in fertilizing peanut or cotton crops [20].

Livestock production is shifting to fewer, much larger operations, categorized as animal feeding operations and CAFOs, with the distinction being the size of the operation (CAFOs house > 1000 animal units) [21]. As a consequence of this increasing intensification of livestock practices, large quantities of manure are consolidated over a limited geographic area, exceeding the demand from nearby farms. The resultant excess manure can pose potential environmental risks when stored or applied in heavier quantities that include high dissolved oxygen levels and increased nitrate concentrations that lead to algal blooms in surface waters [22]. Certain constituents can also cause health problems for grazing animals [23].

The response to these risks has led to more regulation and conservation programs by federal, state, and local governments. In some

Table 1
Summary of national average annual waste feedstock resource estimates for wastewater sludge, animal manure, food waste, and FOG in units of Tg/y and MT/y.

Feedstock	Potential waste resource Tg/y (MT/y)	Category totals Tg/y (MT/y)
Wastewater sludge		
Primary sludge (dry-weight)	7.43 (8.19)	
Secondary sludge (dry-weight)	5.13 (5.65)	
Subtotal		12.56 (13.84)
Animal manure		
Fattened cattle (recoverable, dry-weight)	9.75 (10.75)	
Dairy cow (recoverable, dry-weight)	18.94 (20.87)	
Market hogs (recoverable, dry-weight)	8.97 (9.88)	
Subtotal		*37.66* *(41.50)*
Food waste		
Residential (dry-weight)	8.82 (9.72)	
Commercial (dry-weight)	3.30 (3.64)	
Institutional (dry-weight)	1.37 (1.52)	
Industrial (dry-weight)	0.27 (0.30)	
Subtotal		*13.76* *(15.18)*
Fats, oils, grease (FOG)		
Yellow grease	0.99 (1.10)	
Brown grease	1.50 (1.65)	
Poultry fats	0.75 (0.83)	
Livestock fats	2.13 (2.34)	
Subtotal		*5.37* (5.92)
Total		69.35 (76.44)

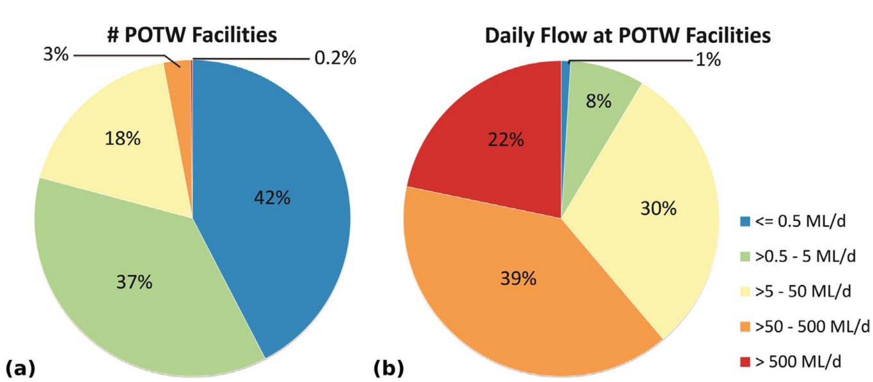

Fig. 2. Fraction of (a) number of POTW facilities, and (b) daily flow, of 15,014 POTWs distributed by facility size (adapted from Seiple et al. [1]).

cases, state and local governments have claimed damages to water resources from manure use, leading to filing of lawsuits against livestock operations. Programs to comply with new regulations increase the cost of livestock operations. Many operations are now required to develop and comply with detailed manure management plans to limit the potential for catastrophic spills or exceedance of the agronomic needs of nearby crops. Alternatives include expanding agreements with farmers to accept manure, acquisition of additional land for manure application, reduction of manure nutrient content, reduction in manure production, and/or finding alternative uses for the manure [20].

One such alternative use of manure is as a renewable energy feedstock. While alternative use in the United States is somewhat limited, the EPA [24] inventoried 259 operational on-farm anaerobic digestion systems for methane gas production from beef, dairy, poultry, and hog operations that provide heat and/or electricity. Though the economics of existing approaches have generally not proved to be attractive, broader potential societal, environmental, and sustainability benefits of using manure as an energy feedstock have been recognized. In particular, in areas where there is an excess of manure and the cost of acquisition is lowest, manure for energy could prove to be a recognized synergistic economic and beneficial use, converting manure from a liability to a revenue generating stream [25].

As depicted in Fig. 3, the primary functions associated with manure handling are production, collection, transfer for storage and/or treatment, and utilization [23]. Production relates to the manure type and amount generated. Collection and storage refer to the "harvesting" of the manure from locations of production and its temporary containment prior to treatment and/or use. Treatment relates to physical, chemical, or biological modification of the manure to reduce pollution potential. Transfer refers to the movement of the manure throughout the management system, and can occur in a solid, liquid, or slurry state. To emphasize the characteristics of manure relevant to HTL processing, we are focused on properties of the manure as it is produced by the animal. This is likely to change in the future as mixing manure with bedding materials, spilled feed, milking house waste, etc., is considered, for example, to represent more realistic conditions of collection, improve biocrude oil quality, and/or achieve a proper solids concentration in an HTL slurry feed.

For this study, estimates of total recoverable manure, or the fraction of total excreted manure likely to be collected from confined livestock operations, generated by cattle, dairy cows, and swine [2] were used to estimate biocrude oil potential. A breakdown and summary of the national estimates of annual average recoverable dry-weight manure production for each livestock category are presented in Table 1.

2.3. Food waste

Two conceptual models discussed here can be used to evaluate

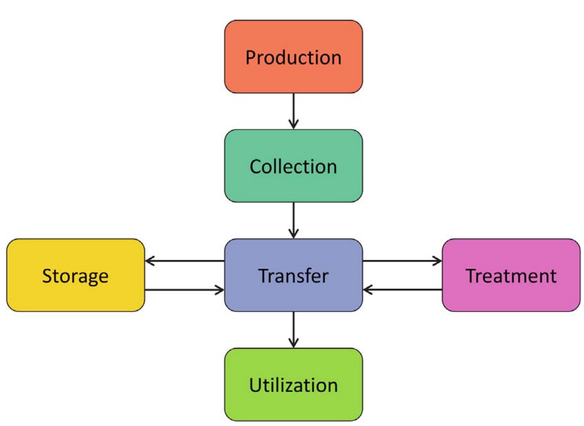

Fig. 3. Typical animal manure handling functions (adapted from USDA [23]).

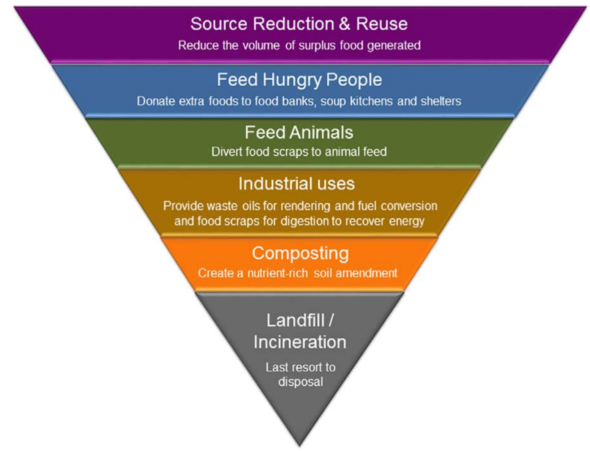

Fig. 4. Food Waste Recovery Hierarchy (adapted from BSR [26], EPA [27]).

opportunities for diversion of food waste as a feedstock for HTL conversion to a biocrude oil. The first is the "Food Value Chain" that encompasses the linkages from "post-agriculture to fork." The main sectors in the value chain outlined in a study by Business for Social Responsibility [26] include 1) industrial, e.g., food processing and manufacturing centers; 2) institutional, e.g., hospitals, universities/schools, prisons; 3) commercial, e.g., grocery stores, restaurants, warehouse/distribution centers; and 4) residential, e.g., households.

The second conceptual model is EPA's Food Waste Recovery Hierarchy depicted in Fig. 4 [26,27] which follows a pattern similar to the overall waste management hierarchy presented earlier (Fig. 1). The top two tiers do not present opportunities for WtE, but, "Tier 3–Feed Animals", provides potential opportunity, depending upon the value of biocrude oil that might be produced and whether there are excess supplies that either are not suitable or exceed the demand for animal feed. The bottom three tiers all represent significant potential for diversion to WtE. There is a correlation of increased viability for energy use moving down the hierarchy, given the combination of the decreasing value of the competing uses and the likelihood of centralized collection. A key aspect inherent to feedstocks derived from food waste is heterogeneous waste mixtures from numerous and diverse sources, and associated handling processes required to achieve economical sustainability and acceptable composition for HTL conversion.

For this study, following the model of Business for Social Responsibility [26], we used the results of Skaggs et al. [2] wherein industrial, institutional, and commercial (IIC) and residential food waste estimates are treated separately. IIC food waste estimates are based on 1) the number of businesses and employees from the 2012 County Business Patterns [28]; 2) institutional data (e.g., hospitals, educational and correctional facilities), and the Homeland Security Infrastructure Program [29]; and 3) a ratio of food disposed per employee or number of beds/students/inmates. Residential food waste estimates are based on per capita food waste generation ranging from 0.03 to 0.21 Mg/person/y. A literature review identified a clustering around 0.12 Mg/person/y [30,31]. In aggregate, Skaggs et al. [2] estimated that 13.76 Tg/y (15.18 MT/y) of food waste was generated annually at IIC facilities and residential entities in the United States and that the residential sector comprised 66% of the total. A categorical breakdown and summary of the national food waste resource is presented in Table 1.

2.4. Fats, oils, and grease

FOG includes yellow grease (refined and used cooking oil), brown grease (trap/interceptor grease), and animal fats (edible/inedible tallow, choice white grease, lard, and poultry fat). Significant quantities of yellow grease are captured in grease traps associated with restaurants, cafeterias, and other large institutional kitchens. Generally, trap grease is collected by independent renderers, disposed of (anaerobic digestion, landfill, or composting), taken to plants for processing into biodiesel, or used as additives for animal feed, soap, or compost [32]. At the household level, waste fat and oils are typically disposed of through the residential sewer or put in the household garbage. Given the high lipid content of FOG, its use directly provides a high conversion efficiency for HTL biocrude oil, but also can be considered as a feedstock blend component to increase the conversion efficiency of other feedstocks or a direct-use fuel or fuel blend.

For yellow and brown grease, resource estimates by Skaggs et al. [2] are based on urban population numbers and grease generation per capita derived from Wiltsee [33]. For inedible animal fats, they used state-level animal (cattle/calves, hogs, chickens, and turkeys) slaughter data from the U.S. Department of Agriculture (USDA) [34]. The USDA state-level data were disaggregated to site location and county-level aggregation using rendering plants' locations and the number of employees per business from a variety of data sources.

Skaggs et al. [2] estimate the inedible FOG production in 2012 to have been 5.37 Tg/y (5.92 MT/y), with livestock fats contributing nearly 40% of the total. A breakdown of the individual FOG categories and summary of the national resource are presented in Table 1.

2.5. Feedstock characteristics

Limited experimental results were found in the literature characterizing the priority feedstocks investigated in this study; thus, the values presented in Table 2 are intended to be representative characteristics only [6,22,35–43].

For wastewater sludge, several investigators presented analytical results including ultimate analysis, proximate analysis, and biochemical component analysis [6,35,44,46–48]. In many studies, the treatment stages of the sludge samples are not specified and therefore are not included here. The representative higher heating values (HHVs) for both primary and secondary treatment sludge were approximately 20 MJ/kg, and the primary sludge had a slightly higher value. Carbon ranged from approximately 43.6–52.5%. Primary sludge had the highest lipid value (18%), and a lower protein content, though carbohydrate content was similar.

Manure characteristics for fattened cattle and dairy cows are not generally distinct. Carbon and oxygen contents are comparable, and the HHVs and lipid and protein contents are slightly higher for fattened cattle manure. Compared to wastewater sludge, both have relatively low HHVs, which correlates with the lower lipid and protein contents and higher carbohydrate content. Correspondingly, compared to cattle manure, swine manure tends to have slightly higher percentages of carbon, oxygen, and lipids.

For the representative food waste results shown, the percentages of carbon and oxygen were similar to those of primary sludge, while the HHVs were somewhat higher. For FOG, as would be expected, the percentages of carbon and lipids are significantly higher than for the other feedstocks.

3. Waste-to-Energy conversion via hydrothermal liquefaction

HTL is a thermochemical conversion of wet biomass into liquid fuels in a hot, pressurized liquid water environment, where water serves as both solvent and reactant. Under processing conditions of approximately 350 °C and 200 atm, and a biomass feedstock in the desirable range of 20 wt% dry solids, a reaction medium is created such that within minutes, biopolymers are deconstructed and reformed into lower molecular weight biocrude oil components. The energy-dense biocrude oil can then be upgraded to a variety of liquid hydrocarbon fuels by hydrotreating [4,48].

HTL has a number of advantages over other thermochemical conversion methods. High lipid concentrations are not required for effective HTL energy conversion and the need for energy-intensive feedstock drying is potentially reduced or eliminated because a feedstock slurry is used as an input [37]. The high-efficiency chemistry of HTL transforms almost all of the biomass into biocrude oil, which largely self-separates from water as the reaction solution returns to standard conditions [44]. In addition, biomass that is not converted through the process can optionally be returned to the input feedstock slurry, thereby increasing biomass to biocrude oil conversion rates. Further, HTL does not generate significant amounts of sludge or hazardous products of combustion such as NOx [49]. The residuals from HTL are 1) a quantity of carbon dioxide gas; 2) a separated solid stream with precipitated phosphates that are of limited supply; and 3) an aqueous stream that optionally can be treated with catalytic hydrothermal gasification to recover the dissolved organic constituents such as a fuel gas and produce a reusable water stream with ammonia nutrients. Akhtar and Amin [49] note that because of the high temperature and pressure, HTL is insensitive to feedstock particle size, which eliminates the need for feedstock particle size reduction, and subsequent energy inputs, to enhance biomass fragmentation and accessibility to treatment. Also, given the hydrophilic nature of most biomass, methods for creation and pumping of biomass feedstock slurries of 5–35% dry solids are generally well known [4,5]. For WtE, the robustness of HTL offers the potential to convert a wide range of feedstocks that otherwise are managed as waste streams. Likewise, HTL processing of sludge and manures sterilizes the

Table 2
Representative biochemical characteristics for seven waste feedstocks.

	Weight % dry basis										
Feedstock	% C	% H	% N	% O	% S	% VM	HHV (MJ/kg)	% Lipid	% Protein	% Carb	Refs.
Primary sludge	47.8	6.5	3.64	33.6	0.48	82.17	20.7	–	–	–	[6]
	51.5[a]	7.0[a]	4.5[a]	35.5[a]	1.5[a]	65	–	18	24	16	[35]
Secondary sludge	43.6	6.55	7.9	29.0	0.72	76.25	19.6	–	–	–	[6]
	52.5[a]	6.0[a]	7.5[a]	33.0[a]	1.0[a]	67	–	8	36	17	[35]
Fattened cattle	35.38[a]	3.73[b]	2.38[b]	57.51[b]	–	16.21[b]	15.16[b]	–	–	–	[22]
	–	–	–	–	–	–	–	6.8	26.6	52.5	[36]
Dairy cows	38.8	5.1	1.3	54.7	–	83.2	11.9	5	18.11	52.6	[37,38]
Market hogs	41.1	5.42	3.36	50.1	–	83.7	–	20.3	24.5	34.7	[39]
Food waste[c]	44.75	6.26	3.36	35.06	0.71	21.0	–	–	–	–	[40]
[b]	–	–	–	–	–	–	36	5.3	18.4	55	[41]
FOG	–	–	–	–	–	96.5	–	78[a]	7[a]	15[a]	[42]
[d]	76.3	11.8	< 0.5	11.5	0.003	–	–	–	–	–	[43]

[a] Presented as % volatile matter (VM).
[b] Wet basis with 76.37% water content.
[c] Food blend.
[d] Rendered animal fat.

bioactive contaminants that are present, thereby reducing environmental risks [5].

4. HTL feedstock conversion and biocrude oil yield

The selection of WtE feedstocks to maximize biocrude oil yield and quality for a given location will depend on a number of factors, including regional availability, feedstock composition, blending potentials, and the economic constraints of feedstock preparation and transport logistics. It is also understood that reaction conditions significantly affect HTL conversion efficiency including reaction temperature, retention time, solids loading, etc., and it is not straightforward to compare yields for different feedstocks and/or different conversion conditions. Nonetheless, a number of investigations have demonstrated that feedstock biochemical composition significantly affects HTL biocrude yield and chemistry, even when bulk elemental profiles are similar [41,49]. These links emphasize the need for feedstock characterization and selection to achieve intended downstream targets [44].

A generally observed trend in the HTL conversion efficiency of biochemical components is the following: lipids > protein > carbohydrates. The low conversion efficiency for carbohydrates is attributed to their higher lignin content, which proves difficult to degrade and mostly ends up as residue, while the major components in the proteins, hemicelluloses and cellulose, are favorable for biocrude oil yield [50]. Akhtar and Amin [49] point out that heterogeneity in biomass feedstocks, particularly related to lignin, hemicelluloses, and cellulose, leads to different behavior under given hydrothermal conditions. One study investigated the relative importance of HTL operating conditions and feedstock composition, comparing yields under two operating temperatures (300 °C for 20 min and 350 °C for 60 min) using model compounds for lipids (sunflower oil and castor oil), proteins (soy proteins and albumin), and polysaccharides (corn starch and cellulose). The results showed that lipids and proteins produce approximately the same yields under both conditions produced, while yields from the polysaccharides are nearly twice as high under the higher temperature conditions. It was also found that the yields for the two polysaccharides were equivalent at both temperatures [51]. Another experiment, using HTL to convert the algae *Scenedesmus* sp., produced biocrude oil yield four times the original lipid content of the biomass, again demonstrating that biocrude oil yield is not just dependent on lipid content [52]. Additional experiments also confirmed that algal biomass richer in lipids and proteins produces higher yields than carbohydrate rich biomass [52]. In conversion experiments using model components of proteins and polysaccharides, it was shown that a mixture of the two materials produces higher yields than protein alone, leading to the trend, mixture > proteins > carbohydrates [51].

For each of the feedstocks, examples of HTL biocrude oil are described in the literature, though the information provided varies from study to study. Example experimental results from some of the HTL conversion experiments are shown in Table 3 for comparison purposes [6,22,37,44–46]. As general observations, biocrude oil yields for primary and secondary wastewater sludge are 37.3% and 24.8%, respectively, with corresponding HHVs of 37.8 MJ/kg and 34.8 MJ/kg [6]. Biocrude oil yields for manure are presented by Yin et al. [22], Midgett et al. [37], Chen et al. [39], Vardon et al. [44], and Xiu et al. [53], and, for example, the conversion yield was 48.8% for fattened cattle and 30.2% for market hogs. Available results for food waste HTL conversion were limited to an undefined blend of food materials and specific food components (e.g., starch and crude protein) [41,45,51,54]. We show the HTL conversion value for the food waste blend, including a 45% biocrude oil yield. No examples of FOG conversion via HTL biocrude oil were found in the literature, but examples for various cooking oils (e.g., vegetable oil and sunflower oil) were found [37,41]. The values for sunflower oil are in Table 3, including the biocrude oil yield of > 90%. Table 3 also shows equivalent values for biodiesel

Table 3
Properties of HTL biocrude oil obtained from WtE feedstocks and reference HHVs for biodiesel.

Feedstock	Biocrude oil yield %	Weight % dry basis					HHV (MJ/kg)	Ref.
		C	H	N	O	S		
Primary	37.3	66.6	10.3	3.7	12.4	0.18	38.8	[6]
Secondary	24.8	71.4	8.68	5.02	7.32	0.89	34.8	[6]
Fattened cattle	48.8	73.7	8.1	1.4	16.8	–	35.5	[22]
Dairy cows	–	–	–	–	–	–	33.5	[37]
Market hogs	30.2	71.2	9.5	3.7	15.6	–	34.7	[44]
Food waste[a]	45	75.2	10.3	3.2	11.3	–	35.1	[45]
FOG[b]	> 90	80.3	11.5	0	8.1	0	42.4	[51]
Biodiesel	N/A	77	12	0	11	N/A	40.15	[22]

[a] Blend.
[b] Sunflower oil.

[6,22,37,44,45,51]. While direct comparison of these results can be difficult due to varying process conditions (e.g., reaction temperatures, retention time, solids loading, etc.), some general observations can be made. Not surprisingly, the highest conversion yield—greater than 90%—is for the representative FOG. For the other feedstocks, yields varied from 48.8% for cattle to 24.8% for secondary sludge. The converted feedstock HHVs varied from 33.5 MJ/kg for dairy manure to 42.4 MJ/kg for FOG which is in the range of biodiesel (40.15 MJ/kg). With the exception of primary sludge, the percentages of carbon for all the feedstocks are > 70%, generally comparable to that of biodiesel.

5. WtE biocrude oil production potential

Our previous efforts to estimate the biocrude oil production potential associated with HTL conversion relied upon a simple regression (Eq. (1)) as follows [58–60]:

$$Y = B^*(0.5638^*l + 0.2106) \quad (1)$$

where Y is biocrude oil yield, B is ash-free dry-weight algae biomass, and l is the lipid fraction of the algae.

This study used and added functionality to Pacific Northwest National Laboratory's Biomass Assessment Tool to estimate the national-scale raw feedstock and biocrude oil production potential from waste feedstocks. The Biomass Assessment Tool is an integrated spatial and numerical modeling suite, analysis platform, and data management architecture that captures site environmental conditions, feedstock production potential, resource requirements, sustainability metrics, and partial techno-economics for a wide range of bioenergy feedstocks [59,61–63]. Part of the objective of the work presented here is to enhance the Biomass Assessment Tool's HTL conversion model to more explicitly account for differences in site-specific feedstock WtE biochemical characteristics and their effect on biocrude oil yield. A critical requirement for such a model is that it is based on readily available data and information for each of the primary feedstocks.

5.1. Reduced-form HTL conversion model

Three general modeling approaches were identified from the literature. In increasing complexity, they include a biochemical composition model [50], a multiple-linear regression model [36], and a quantitative kinetic and reaction network model [52,64]. The basis for the biochemical composition model is the assumption that biocrude oil yield correlates with the feedstock fractions of lipids, proteins, and carbohydrates, and that the behavior of each component is additive [50]. Based on the above assumptions, the simple formula (Eq. (2)) is:

$$B = (L_y \bullet L_c) + (P_y \bullet P_c) + (C_y \bullet C_c) \quad (2)$$

Where B is percent biocrude oil yield, L_y is a lipid yield coefficient, and L_c is percent lipid content, P_y is a protein yield coefficient, P_c is percent protein content, C_y is a carbohydrate yield coefficient, and C_c is percent carbohydrate content. Biller and Ross [50] determined the biocrude oil yields of seven model compounds, including albumin, soya protein, asparagine, glutamine, glucose, starch, and sunflower oil, using HTL with an unstirred bomb type reactor operated at 350 °C for 1 h. Results showed that oil formation was consistent with the trend of lipids > proteins > carbohydrates with biocrude oil yields of 80–55%, 18–11%, and 15–6%, respectively. As an illustration, the authors created a model mixture containing 28.7% lipid fractions (dry), 43% protein, and 27.6% carbohydrate. The mixture was processed by HTL and a biocrude oil yield of 30.1% was observed. The predicted yield, using the above model and the midpoints of the measured ranges from the model compounds, is 28.5%. The difference of 1.6% yield is within acceptable error margin.

An alternative approach to predicting biocrude oil yield based on feedstock biochemical composition is presented by Wang [36]. In this study, HTL tests were conducted on a diverse set of 17 feedstocks including swine manure, cattle manure, sewage sludge, sawdust, multiple types of algae, and various feedstock mixtures. The compositions of the various feedstocks are listed in Table 4 [44,55–57]. Wang [36] established linear regressions to define the relationships between feedstock biochemical components and biocrude oil yield, where correlations were established individually for lipids, proteins, and carbohydrates and resulted in prediction errors from − 0.6% to 14.74% with an average absolute error of 5.3%. The regression equations are presented in Eqs. (3)–(11).

This family of regressions is applied where lipid, protein, and carbohydrate components are evaluated independently and the regression equation used is dependent on the biochemical weight fractions. To determine the fraction of HTL-processed biocrude oil from feedstocks that exhibit a balanced distribution of lipids (X_L), proteins (X_P), and bulk carbohydrates (X_C) (i.e., $X_P < 30$, $X_L < 20$, $X_C < 80$),

$$Y1 = (Y11+Y12+Y13)/3 \tag{3}$$

where

$$Y11 = 1.6701\bullet X_L+8.8709, \tag{4}$$

$$Y12 = 1.1828\bullet X_P+3.5485, \text{ and} \tag{5}$$

$$Y13 = -0.7014\bullet X_C+71.543 \tag{6}$$

Table 4
Biochemical compositions for 17 feedstocks and feedstock combinations (adapted from Wang [36]).

	Weight % of total solids content		
Feedstock	**Lipid**	**Protein**	**Carbohydrate**
Fresh swine manure #1	18.8	26.9	42.9
Nursery swine manure	15.9	29.4	40.5
Fresh swine manure #2 [55]	16.4	25.3	44.1
Manure – soluble fraction in fresh swine manure [55]	29.1	46.2	15.7
Manure – solid fraction in fresh swine manure [55]	5.0	11.4	71.7
Sawdust	0.8	1.5	88.7
Fresh sow manure	20.0	22.6	32.5
Fresh cattle manure	6.8	26.6	52.5
14d pit swine manure + sawdust #1	7.0	15.6	66.2
14d pit swine manure + sawdust #2	10.2	23.1	56.0
Mixture of model compounds #1	15.0	30.0	45.0
Mixture of model compounds #2	20.0	50.0	20.0
Diatom (algae)	5.6	40.0	18.8
Chlorella (algae) [56]	0.5	71.3	23.0
Spirulina (algae) [44]	5.1	64.4	21.0
Algae from Algaewheel [57]	3.7	29.0	29.9
Sewage sludge	0	41.6	41.1

When protein content is high and greater than the lipids fraction, i.e., $X_P > 30$ and $X_L < 30$,

$$Y2 = (Y21+Y22+Y23)/3 \tag{7}$$

where

$$Y21 = 0.0333\bullet X_L+33.565, \tag{8}$$

$$Y22 = 0.1341\bullet X_P+27.059, \text{ and} \tag{9}$$

$$Y23 = -0.0984\bullet X_C+37.114 \tag{10}$$

When carbohydrates dominate the feedstock composition, i.e., $X_C > 80$,

$$Y3 = Y13 \tag{11}$$

Similar to the work by Wang [36], Teri et al. [51] subjected a set of model compounds to HTL processing under different temperature and duration conditions using single and binary/ternary mixed feedstocks. The compounds used included cornstarch and cellulose as model polysaccharides, soy protein and albumin as model proteins, and sunflower oil and castor oil as model lipids. The compounds were treated by HTL at 300 °C and 350 °C for batch holding times of 10–90 min. Teri et al. [51] constructed a regression model to predict biocrude yield that assumed the lipids, polysaccharides, and proteins react independently and interact with one another during HTL having the general form,

$$B = (aX_l + bX_c + cX_p + dX_lX_c + eX_lX_p + fX_cX_p) \tag{12}$$

where B = biocrude oil yield wt%; the X_i terms are the mass fractions of lipid (l), protein (p), carbohydrate (c) in the mixture; and a, b, c, d, e, and f are parameters. The first three terms represent the biocrude oil yield from each of the model compounds alone, while the final three terms account for potential interactions between the components. The measured values of biocrude yield for each of the compounds were used to determine the compound interaction coefficients (terms d, e,and f). The approach assumes a linear relationship between dependent and independent variables and requires a normal data distribution. Interestingly, tests conducted by Teri et al. [51] demonstrated that model accuracy was greatest when the interactive terms were excluded and they suggest the use of a simple linear combination of the individual compound yields.

A third modeling approach, presented by Valdez and Savage [64], Valdez et al. [52], and Hietala and Savage [65], is a reaction network model based on lumped product fractions including gas, solids, and various liquid products as a function of HTL temperature and time. The model was initially developed and demonstrated to describe the HTL processing of the microalgae, *Nannochloropsis* sp., and was subsequently expanded be a more generalized kinetics model for HTL of any algae species under any conditions. Similar to Biller and Ross [50], the model incorporates the biochemical content of the algae such that it can be used for any algae for which the protein, lipid, and carbohydrate content are known.

Fig. 5 illustrates the reaction network for each pathway in the HTL model based on the biochemical content [52]. The model consists of a set of first-order reactions for each pathway that maintains batch-reactor mass balance.

The subscripts for the rate constants (k_j) in Fig. 5 refer to the specific lumped species (e.g., k_3 and k_6 are biocrude to aqueous and gas phases, respectively, and l, p, and c represent lipid, protein, and carbohydrate, respectively). The model assumes that the ash-free protein, lipid, and carbohydrate weight fractions react independently and with the same kinetics, regardless of the algae species being analyzed. A series of six differential equations are solved for each reaction pathway (proteins, lipids, carbohydrates, aqueous phase, biocrude, and gas), while it simultaneously estimates the values of the rate constants (k_j) for a given temperature by minimizing the least square error between observed and predicted values for each product fraction yield at each reaction time for each of the biochemical components. The authors report that

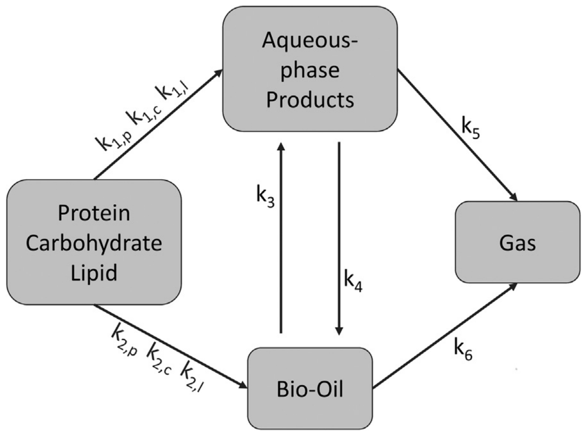

Fig. 5. HTL reaction network incorporating biochemical content from (adapted from Valdez et al. [52]).

for a series of tests with three different algae species, the average standard deviation for data generated at 350 °C was 3.1 wt%.

For the purposes of estimating the biocrude oil production potential from selected WtE feedstocks, the model by Wang [36] was selected for this study. First, given its structure, the model provides robustness by being applicable to a wide range of biochemical compositions. Secondly, as additional HTL feedstock conversion experiment data are generated, they potentially will enable further development and improvement of the model.

6. Results

For this analysis, we used the linear regression approach of Wang [36] and the literature-derived biocomposition values (Table 2) for three of the four waste resource feedstock groups (manure, wastewater sludge, food waste) to develop HTL-based biocrude oil estimates for the conterminous U.S. In the case of FOG, with its high lipid content, the regressions of Wang [36] did not capture a reasonable conversion value, and therefore a fixed value of 0.8 wt biomass/wt biocrude oil was used, which conservatively approaches oil conversions found for pure oils such as sunflower and castor oil [51]. For manure, regression values were established for fattened cattle, dairy cow, and swine manure, but because the range of uncertainties in the biocomposition values is high, individual manure results were averaged to result in a conversion value of 0.32 wt biomass/wt biocrude oil. Additionally, combined primary and secondary wastewater sludge was determined to have a 0.39 wt biomass/wt biocrude oil conversion, and the food waste conversion rate was 0.25. In all cases, to convert from a biocrude oil weight to volume, we assumed a nominal oil density of 0.98 kg/L. Evaluating the unconstrained waste feedstocks converted to biocrude oil, manure is the dominant resource at 45%, followed-by FOG at 20%, wastewater sludge at 19%, and food waste at 16% (Fig. 6).

The biocrude oil conversion results for each feedstock are presented in Table 5 and show the average annual totals in gigaliters (GL) and billions of gallons (Bgal). With the exception of FOG, HTL conversion values (Table 5) were established using average biochemical composition values found in the literature and the Wang [36] regression model. FOG conversion values were set at a fixed 80% for animal fats, yellow grease, and brown grease, which is a more conservative estimate based on sunflower oil conversion values (Table 3). Although the biochemical composition of food waste may vary depending on its source (i.e., residential, commercial, institutional, industrial), using the reduced-form HTL model, we determined a fixed conversion value of 25%, which is much more conservative than the 40% conversion value determined by Zastrow et al. [45]. For the conterminous U.S., the total unconstrained potential biocrude oil resource is estimated to be 22.33 GL/y (5.9 Bgal/y). To provide context for the reported values, Table 5 also shows how

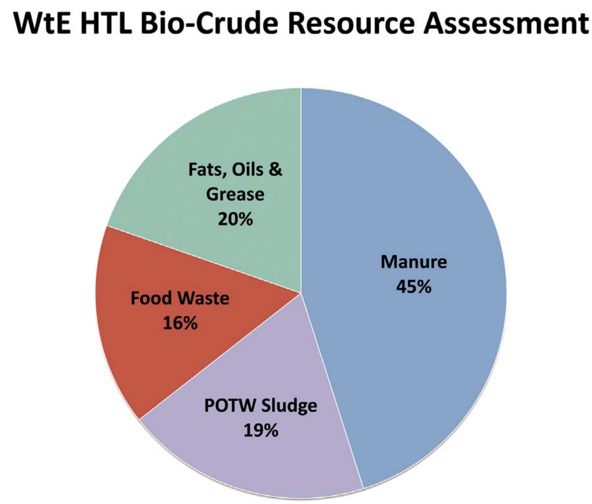

Fig. 6. Total fractions of unconstrained resource availability for major waste feedstocks groups converted to bio-crude oil.

Table 5
The reduced-form HTL model determined feedstock to biocrude oil conversion value, the estimated annual biocrude oil production per waste feedstock for the conterminous U.S., and the percentage of 2016 aviation kerosene demand that could have been met by waste feedstock-sourced biocrude oil.

Feedstock	Reduced-form HTL model conversion value	HTL biocrude oil GL/y (Bg/y)	% of 2016 produced kerosene-type jet fuel
Wastewater sludge			
POTW – primary sludge	0.44	2.82 (0.74)	3.0%
POTW – secondary sludge	0.34	1.50 (0.40)	1.6%
POTW sludge total	–	**4.32 (1.14)**	*4.6%*
Animal manure			
Fattened cattle manure	0.30	2.80 (0.74)	3.0%
Dairy cow manure	0.26	3.78 (1.00)	4.0%
Swine manure	0.41	3.49 (0.92)	3.7%
Manure total	–	**10.07 (2.66)**	*10.8%*
Food waste			
Residential	0.25	2.27 (0.60)	2.4%
Commercial	0.25	0.85 (0.22)	0.9%
Institutional	0.25	0.35 (0.09)	0.4%
Industrial	0.25	0.07 (0.01)	0.1%
Food waste total	–	3.54 (0.92)	*3.8%*
Fats, oils, grease (FOG)			
Animal fats (livestock + poultry)	0.80	2.35 (0.62)	2.5%
Brown grease	0.80	1.23 (0.32)	1.3%
Yellow grease	0.80	0.81 (0.21)	0.9%
FOG total	0.80	**4.39** (1.15)	*4.7%*
All feedstock total	–	**22.32 (5.87)**	23.9%

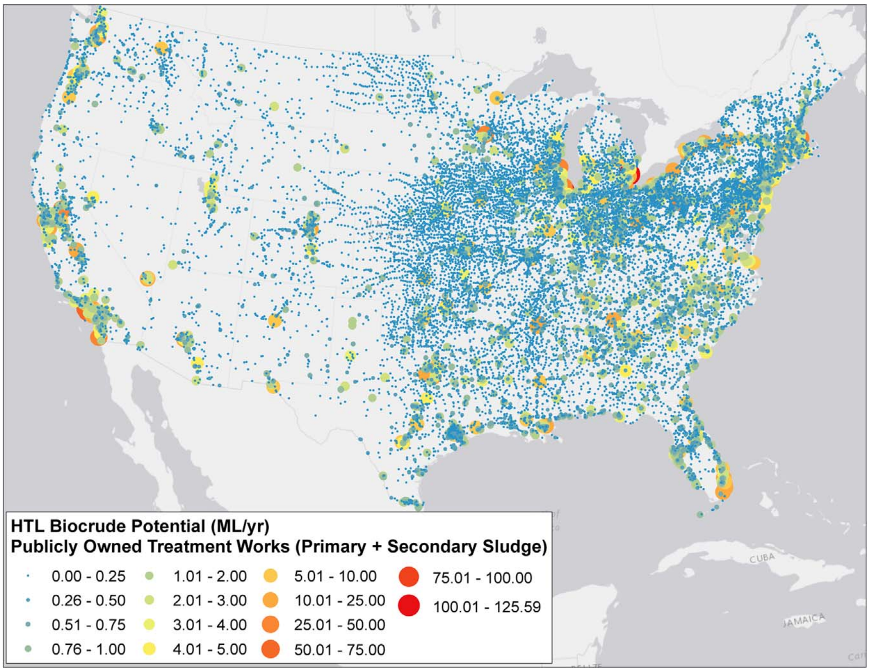

Fig. 7. HTL-processed bio-crude oil potential from combined primary and secondary wastewater sludge from publicly owned treatment works.

each feedstock could contribute to meeting kerosene-type jet fuel (most common) demand. In total, the results show that the considered waste feedstocks could potentially meet 23.95% of the 2016 aviation kerosene demand. This datum is based on U.S. total supplied kerosene-type jet fuel in 2016 [66].

Evaluating the results spatially, Fig. 7 presents the biocrude oil resource potential for primary and secondary sludge sourced from POTWs, which can potentially produce 4.3 GL/y (1.1 Bgal/y). As might be expected, this resource is widely distributed throughout the country and follows population density patterns; there is a high-density of sites in the eastern half of the U.S. and larger individual sites are located in the biggest cities. For manure (Fig. 8), the Midwest and Great Lakes region has the highest density of resource availability, and a large number of smaller individual sites exist throughout Ohio, Indiana, Illinois, Iowa, and Missouri, mixed with numerous large CAFOs in Michigan, Wisconsin, South Dakota, Nebraska, Kansas, Oklahoma, and Texas. Concentrations of manure resources in California, Washington, Idaho, Utah, and southern Arizona are significant as well, and North Carolina, Pennsylvania, and New York show resource availability in the eastern United States. In total, the calculated biocrude oil potential for livestock manures is 10.1 GL/y (2.7 Bgal/y). As with wastewater sludge, the potential resource availability for food waste (Fig. 9) follows population patterns; high-density populations such as Los Angeles, Seattle, Las Vegas, Phoenix, Chicago, Houston, Miami, and New York show high resource availability and thus high biocrude oil production potential. In total, food waste is estimated to produce 3.6 GL/y (0.9 Bgal/y), which is substantial given that this feedstock has the lowest biocrude oil conversion rate out of the assessed feedstocks. For

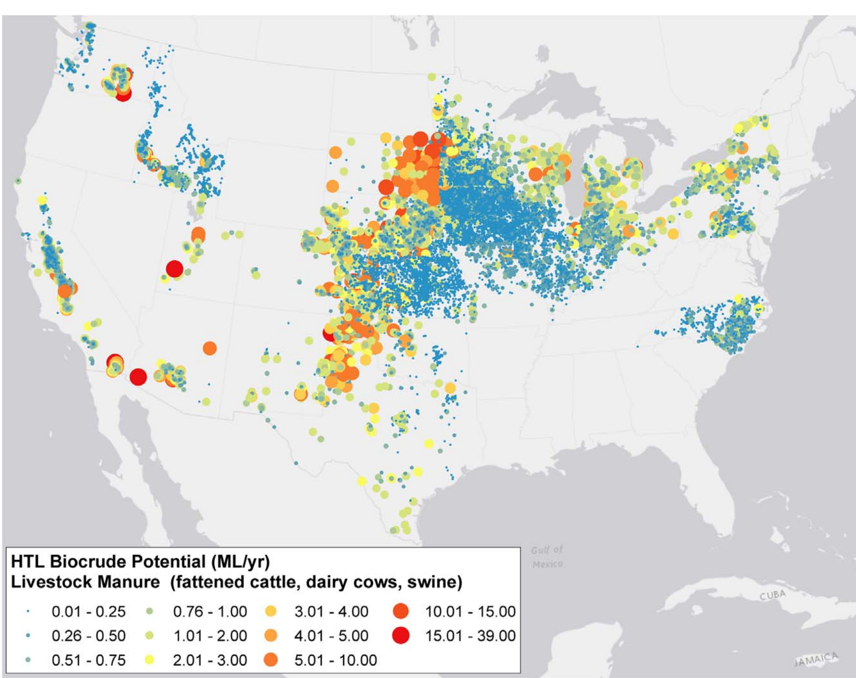

Fig. 8. HTL-processed bio-crude oil potential from livestock manure, including fattened cattle, dairy cows, and swine.

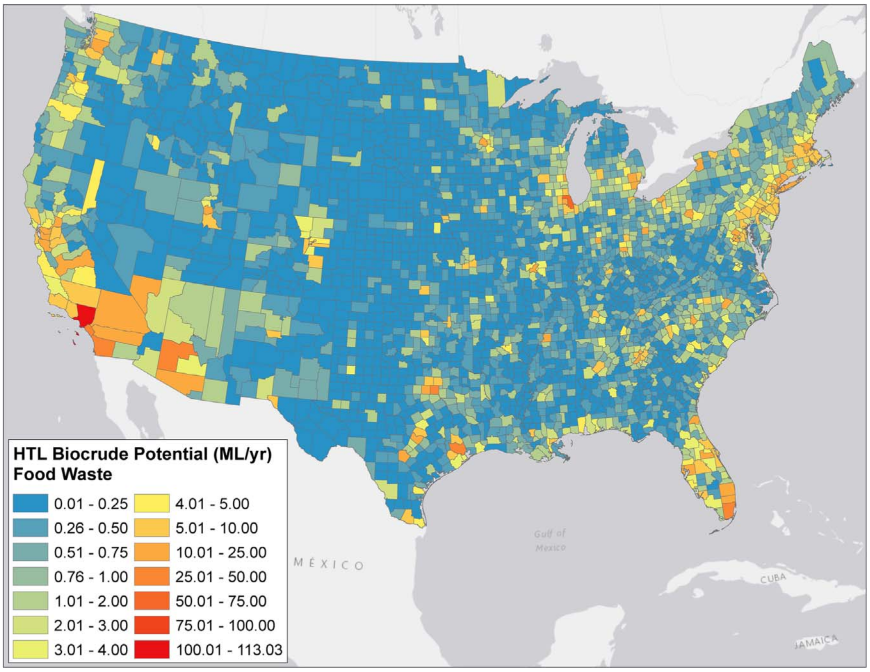

Fig. 9. HTL-processed bio-crude oil potential from county-level estimates of food waste.

FOG (Fig. 10), site resources are reasonably well distributed throughout the United States, with higher density occurring in the eastern half of the country. Several very large facilities are located in the Midwest region and some outlying facilities are in California and New Jersey. With a high HTL conversion rate (0.8 wt/wt), FOG can potentially provide 4.39 GL/y (1.16 Bgal/y) of biocrude oil.

7. Conclusion

The best available data on WtE feedstock resources, including wastewater sludge, animal manure, food waste, and FOG, were coupled with the Biomass Assessment Tool modeling framework and an HTL conversion model to estimate the quantities and geographic distribution of potential biocrude oil production from selected organic wastes. The results of these analyses indicate that for the conterminous United States, on average as much as 22.3 GL/y (5.9 Bgal/y) of biocrude oil could be produced. Future resource assessment efforts need to consider a number of questions not addressed by this preliminary effort. For example, what effect will competing uses have on net feedstock availability for WtE conversion? To what extent can WtE feedstocks be blended on a site-by-site basis to maximize throughput potential and meet feedstock characteristic requirements for conversion technologies such as HTL? What are the major logistical and scaling challenges and tradeoffs for acquiring and preprocessing individual or blended feedstocks for conversion to WtE? And finally, what are the cost-benefit tradeoffs for WtE biocrude relative to conventional fuels?

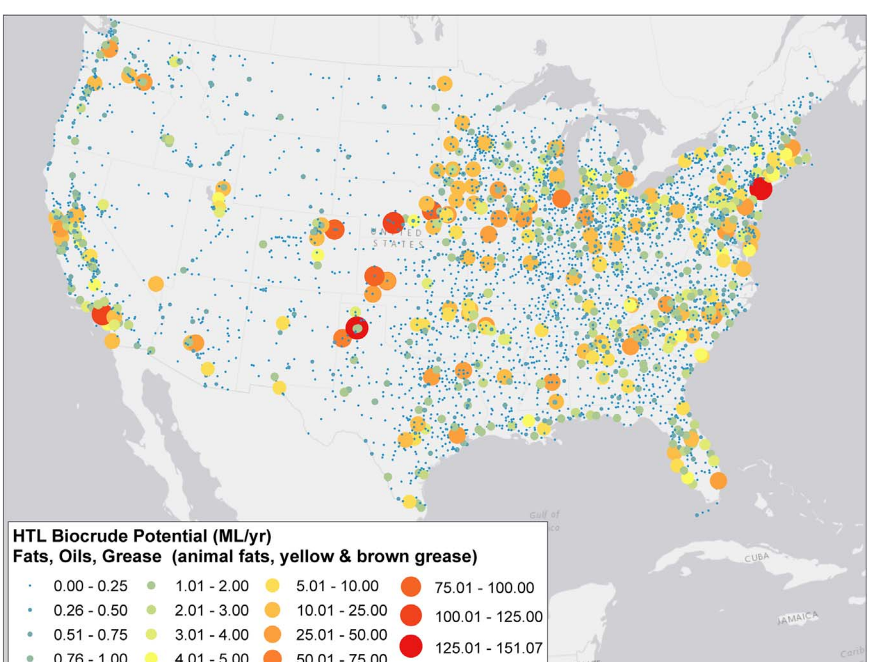

Fig. 10. HTL-processed bio-crude oil potential from fats, oils and grease sourced from animal fats, yellow and brown grease.

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